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Apportionment of Residential Indoor Aerosol, VOC, and Aldehyde Species to Indoor and Outdoor Sources

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The average concentrations of a large number of fine particle aerosol, VOC and aldehyde species measured in ten Boise ID residences in wintertime have been apportioned according to their contributions from all inside sources and all outside sources. The method depends on the availability of average concentrations measured outside each residence during the same sampling periods used for the inside measurements, and on the ability to identify one or more species that have negligible indoor sources.

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APPORTIONMENT OF RESIDENTIAL INDOOR AEROSOL, VOC, AND ALDEHYDE SPECIES TO INDOOR AND OUTDOOR SOURCES

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The average concentrations of a large number of fine particle aerosol, VOC and aldehyde species measured in ten Boise ID residences in wintertime have been apportioned according to their contributions from all inside sources and all outside sources. The method depends on the availability of average concentrations measured outside each residence during the same sampling periods used for the inside measurements, and on the ability to identify one or more species that have negligible indoor sources.

INTRODUCTION

Dockery and Spengler (1) have shown that the average indoor concentration of a stable air pollutant species during a sampling period i, $\langle I \rangle_i$, is related to its average outdoor concentration during i, $\langle O \rangle_i$, by the expression

$$\langle I \rangle_i = m \langle O \rangle_i + b + c \qquad (1)$$

where

$$m = P A / (A + k) \qquad (2)$$

$$b = \langle S \rangle_i / (V (A + k)) \qquad (3)$$

$$c = (I(0) - I(T)) / (T (A + k)) \qquad (4)$$

with P the penetration of the outside pollutant ($0 \leq P \leq 1$), A the air change rate (h^{-1}), k the indoor removal rate (h^{-1}), V the interior volume, T the sampling duration (h), S the time varying rate at which the pollutant is emitted by indoor sources ($\mu g\ h^{-1}$), and I(0) and I(T) the pollutant's indoor concentration at the beginning and end of the sampling period, respectively. The quantity m is also referred to as an infiltration or protection factor, since it is the indoor/outdoor ratio in the absence of indoor sources.

Equations (1) - (4) are based on the assumptions of rapid and uniform mixing of the pollutant throughout the interior volume, a first order removal process, and constant values of A and k. Within these assumptions the equations are exact, valid for any

value of T, and for any arbitrary time variation of O and S. To avoid the complication of eq.(4) however, T should be large enough that term c can be neglected in eq.(1).

The previous equations suggest the following approach for separating the measured indoor concentration of a species into its outdoor and indoor components:

1. Use species with no indoor sources to determine m = <I>/<O> for each residence.

2. For any species, outdoor contribution to indoor concentration = m * (measured outdoor concentration).

3. Indoor contribution to indoor concentration = (measured indoor concentration) - (outdoor contribution to indoor concentration).

This approach has been applied to a variety of fine (0 - 2.5 μm aerodynamic dia) particulate species, volatile organic compounds (VOCs), and aldehydes measured in Boise ID during the 1986-1987 winter under EPA's Integrated Air Cancer Project. In the following, results are given for VOC and aldehyde species not included in a previous report (2) based on the same method and data set.

SAMPLING AND ANALYSIS

Sampling was conducted for a four-day period at each of ten residences, involving a different residence during each successive week between November 15, 1986, and February 4, 1987. Up to eight consecutive 12-h sample pairs of each type were collected simultaneously indoors and outdoors at each residence. Homes with smokers, woodburning appliances and other obvious indoor sources (e.g., kerosene heaters) were deliberately excluded. Description of the sampling and analysis has been given previously for the fine particulate and VOCs (2), and aldehydes (3).

RESULTS

Aerosol Species

A survey of the fine particle data indicated that the species which best satisfied the "no indoor sources" assumption (step 1) were S, Pb, Zn, and K' (soil-corrected potassium, as described previously (4)). The signature for this condition is a zero intercept for a Y-X plot of concentration data measured simultaneously inside and immediately outside a residence. Figure 1 shows such a plot for residence R26. The linear fit using weighted (both coordinates) least squares shows a slope ("m" in eq. (1)) of 0.66 ± .05 and negligible intercept. Similar fits, i.e., negligible intercepts and well-defined slopes less than unity, were found for the other nine residences. Applying steps 1-3 showed that for most homes the indoor source contribution was dominant for fine particle Si, Ca, and Fe, while the infiltration contribution was dominant for S, K, Pb, Zn, mass, and extractable organic matter. Quantitative results have been given previously (2).

Volatile Organic Compounds

The raw data set obtained from the GC analyses included a total of 69 individual species and compound classes. On the bases of data accuracy and completeness, 30 species were selected for detailed consideration. Most of the 30 had no missing or below detection limit measurement values; the remainder had no more than 10 % of their measurements below detection limits. The final data set contained 47 indoor-outdoor measurement pairs for most of the 30 species. Sixteen of the 47 were 12-h sampling periods while the remainder were diurnally-averaged 24-h sampling periods (2).

Figure 2 shows the indoor-outdoor relationship for six VOCs at the same residence (R26) examined in fig. 1 for fine aerosol species. The slope (infiltration factor) in fig. 2 is unity within the uncertainty of the data, which from eq.(2) is only possible if both $P = 1$ and $k = 0$ (more precisely, if k is small in comparison with the air change rate). Eight of the ten residences showed results similar to fig. 2 (unity slope and negligible intercept). The data from the other two may have been influenced by a systematic error (2). Consequently no source apportionment results for those two residences will be reported.

Table 1 gives the results of applying steps 1-3 to 11 of the VOCs, taking the infiltration, m, in step 2 to be unity for all residences. Each entry gives the <u>modeled</u> average indoor or outdoor contribution (ppbC) to the total average indoor concentration measured for that species, separately for each residence, and also averaged over the eight residences shown. The average <u>measured</u> indoor concentration is the sum "OUT + IN". Several of the indoor source values are so large (e.g., n-Hexane in R11) as to raise doubts about data validity. However since virtually none of the outdoor values exhibit any extreme values, there seems no good reason not to accept these cases as valid data. Analogous results are available (2) for 19 additional VOCs.

Aldehydes

In contrast to the VOCs all 12-h aldehyde measurements were used directly, without diurnal averaging. This resulted in a data set of 78 indoor-outdoor measurement pairs for each species. Five of the species had no more than 2 measured values below the limit of detection. An additional species, hexanaldehyde, had 14 such values. These six species were selected for apportionment analysis. Additional species not reported here were measured, but were frequently below the detection limit.

None of the aldehydes exhibited a 1:1 relationship like that shown in fig. 2 for several of the VOCs. Instead the result "indoor > outdoor" was found for virtually every measurement pair. This is consistent with $m = 1$ (i.e., $P = 1$ and $k = 0$) but not conclusive. Although the assumption $m = 1$ has been used in obtaining the aldehyde entries (ppb) in Table 1, the strict interpretation of these values is that each "OUT" is an <u>upper limit</u> for the outdoor contribution to the measured indoor concentration, while each "IN" is a <u>lower limit</u> for the measured indoor concentration due to indoor sources. Because of its adverse health effects the formaldehyde results are noteworthy. Its dominance by inside sources is likely due to direct emissions from wood products (e.g., particle board) rather than photochemical activity.

CONCLUSION

The average concentrations of a large number of fine particle aerosol, VOC and aldehyde species measured in ten Boise ID residences in wintertime have been apportioned according to their contributions from all inside sources and all outside sources. None of the residences had obvious major indoor sources (smokers, woodburning appliances, etc.). The two category apportionment was accomplished through use of the "standard" single chamber mass balance indoor air quality model. The method depends on the availability of average concentrations measured outside each residence during the same sampling periods used for the inside measurements, and on the ability to identify one or more species that have negligible indoor sources. The relative importance of indoor and outdoor sources to measured indoor concentrations showed great variation between species and between residences. In most homes the indoor source contribution was dominant for fine particle Si, Ca, and Fe, while the infiltration contribution was dominant for S, K, Pb, Zn, mass, and extractable organic matter. Indoor contributions to individual VOCs were frequently very large at a few residences and negligible at the others. All aldehydes were dominated by indoor sources.

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Table 1. Average indoor concentrations at each residence from outside (OUT) and inside (IN) sources. VOC units are ppbC. Aldehyde units are ppb.

VOC/ALDEHYDE		R05	R08	R11	R14	R17	R20	R26	R29	ALL
Ethane	OUT	11	14	20	12	11	14	21	13	14
	IN	5	94	77	24	0	0	11	17	25
Propane	OUT	8	9	12	9	10	11	22	13	12
	IN	3	9	7	74	0	3	230	9	43
i-Butane	OUT	5	7	9	5	9	8	20	11	9
	IN	192	4	6	28	0	7	140	9	43
n-Butane	OUT	19	31	43	18	30	28	73	37	34
	IN	9	5	-4	34	3	1	307	13	42
n-Pentane	OUT	11	17	22	10	14	13	32	15	16
	IN	4	1	11	2	3	1	4	6	4
3-Me-Pentane	OUT	7	9	11	6	7	6	16	7	8
	IN	11	1	329	22	12	2	2	10	42
n-Hexane	OUT	9	9	13	7	12	11	24	11	11
	IN	0	2	1935	22	0	0	4	8	205
2,4-Di-Me-Pentane	OUT	3	4	5	2	3	3	7	2	3
	IN	0	0	7	2	7	0	0	4	3
2-Me-Hexane	OUT	8	13	15	7	9	8	22	8	10
	IN	-1	-1	1	5	-2	2	-1	3	2
n-Heptane	OUT	3	5	7	3	6	4	9	4	5
	IN	0	1	4	8	0	4	0	1	3
Chloro-Benzene	OUT	5	3	4	4	7	4	6	5	5
	IN	-1	0	1	2	5	5	19	11	6
Formaldehyde	OUT	3	5	4	6	4	5	7	3	5
	IN	12	6	21	20	5	10	10	19	13
Acetaldehyde	OUT	1	2	2	3	2	2	4	2	2
	IN	13	4	9	8	2	4	14	21	9
Acetone	OUT	1	1	2	2	1	4	2	1	2
	IN	34	12	10	19	4	9	7	9	13
Propion-aldehyde	OUT	0.2	0.3	0.3	0.5	0.3	0.5	0.6	0.4	0.4
	IN	1.4	0.5	0.8	0.6	0.5	0.4	1.0	4.3	1.2
Butyraldehyde + Methylethyl Ketone	OUT	0.3	0.3	0.3	0.6	0.3	0.5	0.5	0.3	0.4
	IN	10.3	0.1	2.5	8.2	0.3	0.5	0.9	3.6	3.3
Hexanaldehyde	OUT	0.0	0.0	0.0	0.4	0.1	0.0	0.3	0.1	0.1
	IN	2.0	0.8	2.4	1.0	0.6	0.7	1.4	2.5	1.4
Total Aldehydes, excluding Acetone	OUT	6	9	8	13	7	10	14	8	9
	IN	45	13	41	41	9	16	29	54	31

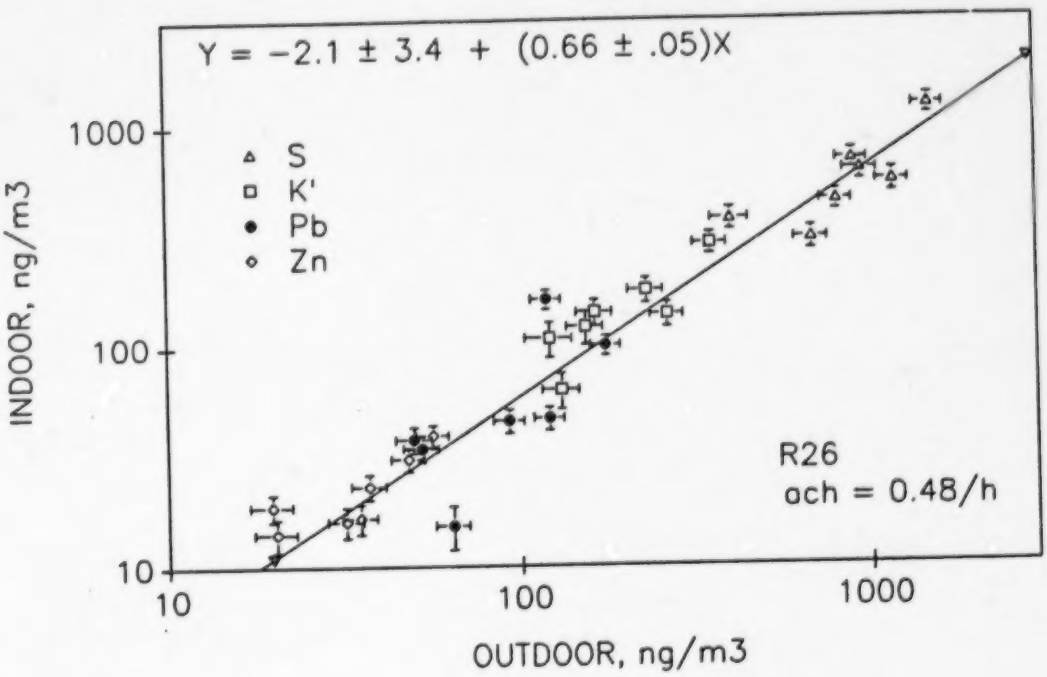

Figure 1. Indoor-outdoor comparison at R26 for fine particle species with small indoor sources.

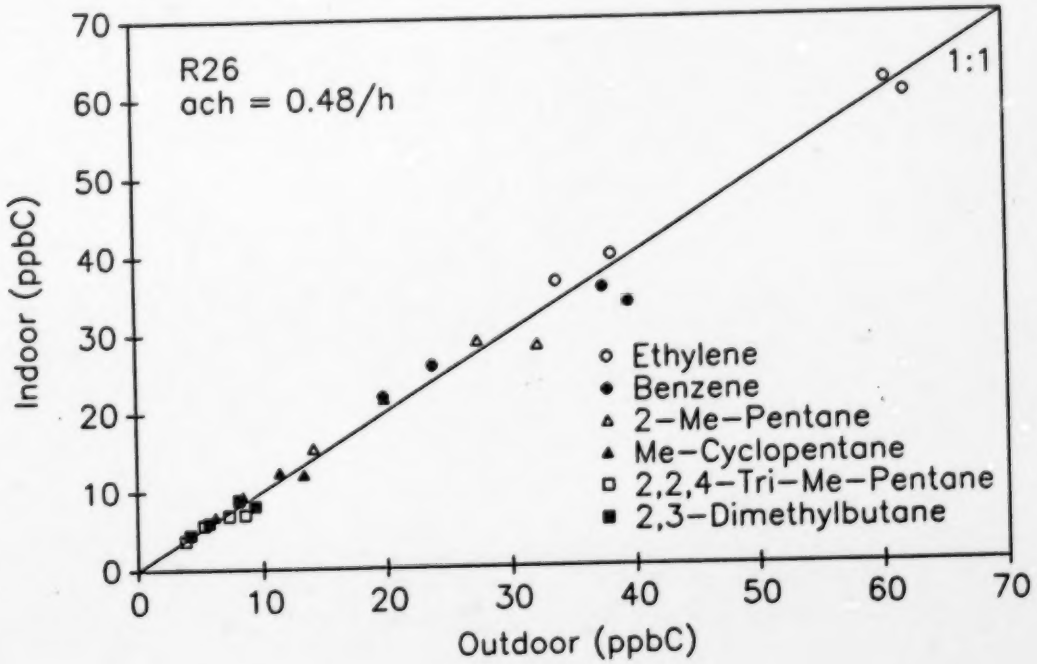

Figure 2. Indoor-outdoor comparison at R26 for VOCs with small indoor sources.